

Antiferromagnetic fluctuations and the Fulde-Ferrell-Larkin-Ovchinnikov state in CeCoIn₅

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The heavy-fermion superconductor CeCoIn₅ is the first material, where different experimental probes show strong evidence pointing to the realization of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state. The inhomogeneous superconducting FFLO state with a periodically modulated order parameter was predicted to appear in Pauli-limited, sufficiently clean type-II superconductors already more than 40 years ago. On the other hand, CeCoIn₅ is supposed to be close to a magnetic quantum critical point (QCP) showing strong antiferromagnetic (AFM) spin fluctuations (SF) at atmospheric pressure. We studied the evolution of the FFLO phase away from the influence of the strong AFM-SF by heat capacity experiments under pressure (0 GPa $\leq P \leq 1.5$ GPa, 0 T $\leq \mu_0 H \leq 14$ T, and 100 mK $\leq T \leq 4$ K). Our results prove the stability of the the FFLO phase under pressure. It even expands, while the Pauli-limiting becomes weaker and the AFM-SF are suppressed. This shows the intriguing influence of the AFM-SF on the FFLO state.

KEYWORDS: superconductivity, high pressure, heavy fermion, FFLO state

1. Introduction

Recently, the possible realization of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting (SC) state in CeCoIn₅ attracted lot of attention.^{1,2} The FFLO state is a spatially inhomogeneous SC phase with a periodically modulated order parameter.^{3,4} It was predicted to appear in clean-limit type-II superconductors close to the upper critical field, $H_{c2}(0)$, if the orbital pair breaking is small relative to the Pauli-limiting effect.⁵ Furthermore, it has been shown that a low-dimensional electronic structure and d -wave symmetry of the SC order parameter reinforces the stability range of the FFLO state.^{6,7} CeCoIn₅ fulfills all these conditions: (i) CeCoIn₅ is in the clean limit;⁸ (ii) the Pauli-limiting exceeds the orbital-limiting effect, indicated by a Maki parameter⁹ $\alpha = \sqrt{2}H_{orb}/H_P > 1.8$ (H_{orb} and H_P are the orbital- and the Pauli-limiting field, respectively);² (iii) the electronic structure is highly anisotropic, dominated by warped cylindrical Fermi-sheets;¹⁰ (iv) superconductivity in CeCoIn₅ is of d -wave type, most likely of $d_{x^2-y^2}$ symmetry, based on results of specific heat,^{8,11} thermal conductivity,^{8,12} NMR relaxation rate,^{13,14} penetration depth,^{15,16} and Andreev-reflection measurements.^{17,18} This makes CeCoIn₅ a favored candidate for the realization of the FFLO state, and indeed different physical properties indicate an anomaly inside the SC state, taken as the transition from the Abrikosov vortex state to the FFLO phase.^{1,2,19-21}

At atmospheric pressure, CeCoIn₅ shows the hallmarks of non-Fermi-liquid (NFL) behavior expected in the vicinity of a magnetic instability. In the normal state close to the upper critical field (i) the Sommerfeld-coefficient, γ , is diverging logarithmically,²²⁻²⁴ (ii) the electrical resistivity ($\rho - \rho_0$) $\propto T$,^{22,24-26} and (iii) the spin-lattice relaxation rate, $1/T_1 \propto T^{1/4}$.^{14,28} These de-

pendencies point to the vicinity of an antiferromagnetic (AFM) quantum critical point (QCP). In detailed studies of electrical resistivity and specific heat as a function of temperature in different magnetic fields ($H > H_{c2}(0)$) the characteristic temperature, T_{FL} , below which Landau-Fermi-liquid (LFL) behavior is recovered appears to vanish at a critical field, H_{QCP} , close to $H_{c2}(0)$ suggesting the existence of a magnetic QCP.²⁴⁻²⁶ Recent experiments show that CeCoIn₅ becomes AFM on slight Cd-doping supporting the proximity to a $H = 0$ AFM-QCP.²⁷ This is corroborated by the comparison of the pressure-temperature (P - T) phase diagrams of CeRhIn₅ and CeCoIn₅, which leads to the conclusion that CeCoIn₅ is close to a AFM-QCP at a small, slightly negative pressure.²⁹⁻³³

The SC FFLO state and the spin fluctuations (SF), possibly AFM, emerging in the vicinity of the field-tuned magnetic QCP close to $H_{c2}(0)$ appear in, or even *share*, only a small part of the H - T phase diagram. The aim of this paper is to study the mutual connection of the AFM-SF and the FFLO state.

2. Experimental

In general, in Ce-based inter metallic compounds the application of hydrostatic pressure suppresses magnetism and eventually a non-magnetic state is achieved at high pressures. In CeCoIn₅ hydrostatic pressure strongly suppresses the AFM-SF. Electrical resistivity³¹ and specific heat³⁴ studies under pressure indicate that the AFM-SF are already significantly suppressed at $P \gtrsim 1.5$ GPa, and LFL is recovered at low temperatures. Additional evidence for the recovery of the LFL state comes from pressure dependent de Haas-van Alphen³⁵ and NMR experiments.¹³ Therefore, heat capacity measurements under pressure, at low temperatures and in magnetic fields are the especially suited to study the evolution of the H - T phase diagram with pressure and to investigate

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the FFLO phase away from the influence of strong magnetic fluctuations.

The heat capacity experiments were carried out on high-quality single crystals of CeCoIn_5 grown from excess In-flux. CeCoIn_5 crystallizes in the tetragonal HoCoGa_5 crystal structure³⁶ that can be viewed as layers of CeIn_3 and CoIn_2 units stacked sequentially along the c -axis.²² A miniature Cu-Be piston-cylinder-type pressure cell was utilized to generate pressures up to 1.5 GPa. In a dilution cryostat, a quasi-adiabatic heat-pulse technique for temperatures down to 100 mK and magnetic fields up to 12 T was employed, while a relaxation method was used in a commercial Physical Property Measurement System (Quantum Design) for magnetic fields up to 14 T and temperatures $0.350 \text{ K} \leq T \leq 4 \text{ K}$. In all experiments the magnetic field was either applied in the ab -plane or parallel to the c -direction of the tetragonal structure. The narrow width of the SC transition of lead which served as pressure gauge/medium confirmed the good quasi-hydrostatic pressure conditions inside the pressure cell. Special care was taken to ensure a precise orientation of the sample with respect to magnetic field H .

Heat capacity measurements were carried out at 0, 0.45, and 1.34 GPa. The highest pressure was chosen in a way that the strong SF present at ambient pressure are already substantially suppressed. In the following we focus our discussion on the pressure evolution of the $H - T$ phase diagram obtained for the magnetic field applied in the ab -plane. For this orientation the existence of the anomaly inside the SC state taken as a signature of the FFLO state has been confirmed by different physical probes.^{1, 2, 19–21} Figure 1 shows specific heat data, $C - C_{\text{Schottky}}$, collected at $\mu_0 H = 12 \text{ T}$ for $P = 0.45 \text{ GPa}$ and $P = 1.34 \text{ GPa}$. The contribution of the nuclear Schottky anomaly to the specific heat at low- T , C_{Schottky} , has been subtracted from the data. Be-

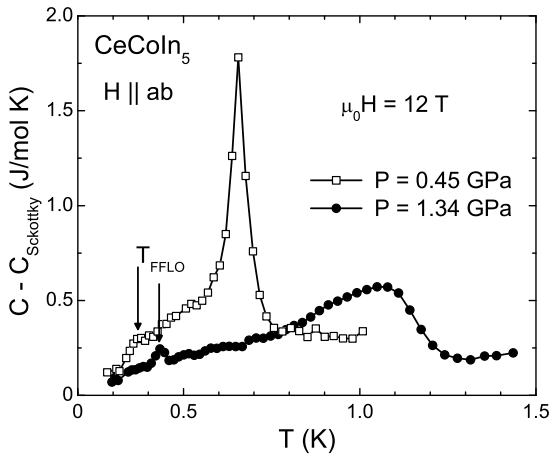


Fig. 1. Specific heat of CeCoIn_5 at $\mu_0 H = 12 \text{ T}$ ($H \parallel ab$) for $P = 0.45 \text{ GPa}$ and $P = 1.34 \text{ GPa}$. The nuclear Schottky contribution to the heat capacity has been subtracted from the data. The low temperature anomaly at T_{FFLO} is indicated by an arrow.

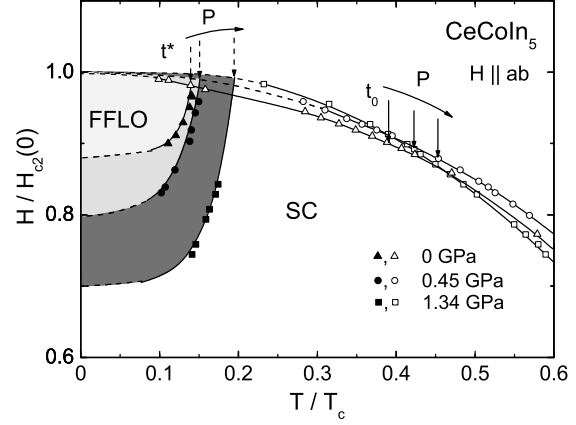


Fig. 2. Combined $H - T$ phase diagram of CeCoIn_5 for magnetic field parallel to the ab -plane for 0, 0.45, and 1.34 GPa. The magnetic field axis and the temperature axis are scaled by $H_{c2}(0)$ and T_c , respectively. Open symbols indicate the transition from the normal to the SC state, while solid symbols mark the anomaly inside the SC state at $t_{\text{FFLO}} = T_{\text{FFLO}}/T_c$. The tricritical point at $t^* = T^*/T_c$, where the FFLO transition line hits the SC phase transition line, is marked by a dashed arrow for each pressure, respectively. The crossover temperature from a second-order to a first-order SC phase transition is indicated by a solid arrow. t^* as well as t_0 shift to higher temperature with increasing pressure.

low the SC transition a second anomaly appears (denoted as $t_{\text{FFLO}} = T_{\text{FFLO}}/T_c$ in Fig. 1) like the one observed at atmospheric pressure. With increasing pressure, T_{FFLO} is moving to higher temperatures. The evolution of the $t_{\text{FFLO}}(P) = T_{\text{FFLO}}(P)/T_c$ phase line in the $H - T$ phase diagram with pressure is depicted in Fig. 2. In this phase diagram H is scaled by the respective $H_{c2}(0)$ and T by T_c for each pressure. With increasing pressure the FFLO phase is expanding under pressure. It appears already at smaller reduced fields, $h = H/H_{c2}(0)$: $h \approx 0.88$ at ambient pressure and $h \approx 0.8$ and $h \approx 0.7$ at 0.45 GPa and 1.34 GPa, respectively. At a constant field, $t_{\text{FFLO}}(h)$ shifts continuously to higher temperature, similar to what is observed for the tricritical point at t^* , where the extrapolated $t_{\text{FFLO}}(h)$ line meets the SC phase transition line. Note, that we did not detect any additional anomaly for magnetic fields above the upper-critical field at any pressure. Besides the second anomaly inside the SC state we observe a remarkable change of both the shape and the size of the anomaly at the SC transition upon increasing the magnetic field. The mean-field-type shape at low fields sharpens up and becomes more symmetrical at high magnetic fields, despite the SC phase transition line being crossed at a glancing angle. This indicates a crossover from a second-order to a first-order phase transition, which we observe at all investigated pressures. The crossover temperature $t_0 = T_0/T_c$ is enhanced from $t_0 = 0.39$ at atmospheric pressure to $t_0 = 0.42$ at 0.45 GPa and $t_0 = 0.45$ at 1.34 GPa, respectively. Our data are in good agreement with results from magnetization studies under pressure.³⁷ At atmospheric

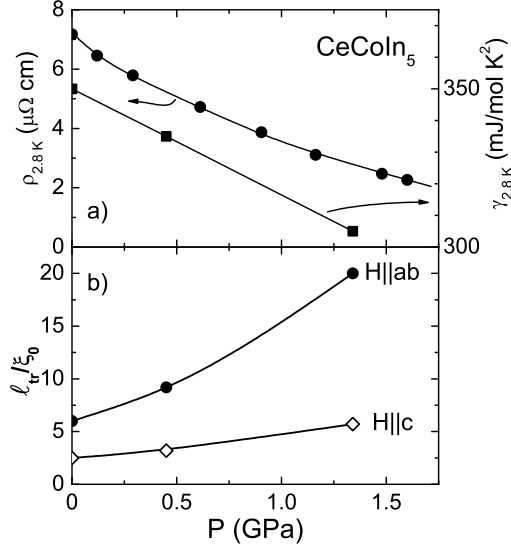


Fig. 3. Upper panel: left axis, resistivity at 2.8 K, $\rho_{2.8K}$ (taken from Nicklas *et al.*³³) and, right axis, Sommerfeld-coefficient at 2.8 K, $\gamma_{2.8K} = C/T(2.8K)$ as function of pressure; lower panel: ratio of estimated mean-free-path, ℓ_{tr} , and coherence length, ξ_0 , for $H \parallel ab$ and $H \parallel c$ as function of pressure. See text for details.

pressure the T_{FFLO} anomaly is only observed above the crossover field $H_0 = H_{c2}(T_0)$ (see also Bianchi *et al.*²). This is not the case under applied pressure anymore. Here, the SC anomaly still displays a mean-field-type shape, indicating a second-order phase transition, while the T_{FFLO} anomaly is already appearing at lower T inside the SC state. The SC transition is of first order only at fields greater than H_0 , much higher than the field where the FFLO anomaly first occurs. Figure 1 shows data at $P = 0.45$ GPa and $\mu_0 H = 12$ T where the transition to the SC state is of first order ($H_0 = 11.2$ T/ μ_0), while for $P = 1.34$ GPa at the same field the transition still displays a mean-field-type shape ($H_0 = 12.4$ T/ μ_0).

3. Discussion

In the following we want to verify, if the necessary conditions for the formation of the FFLO state are still fulfilled in CeCoIn₅ under pressure: i) clean-limit type II superconductor and ii) Pauli-limited with a Maki-Parameter $\alpha > 1.8$. Finally, we will discuss the influence of AFM-SF on the FFLO phase.

The ratio of the quasiparticle mean-free path and the coherence length, ℓ_{tr}/ξ_0 , increases significantly with pressure for the magnetic field applied in the basal plane as well as for the field perpendicular to it (see Fig. 3, lower panel). CeCoIn₅ becomes even *cleaner* with pressure. An upper limit for ξ_0 can be obtained using the BCS relation $\xi_0 = \sqrt{\Phi_0/(2\pi\mu_0 H_{c2}(0))}$, where Φ_0 is the flux quantum. For the estimation of ℓ_{tr} we follow the scheme of Orlando *et al.*,³⁸ where the quasiparticle mean-free path is given by $\ell_{tr} \propto 1/(\xi_0 T_c \gamma_n \rho_n)$. Here, γ_n and ρ_n are the Sommerfeld-coefficient and the resistivity in the normal state right above the SC transition, respec-

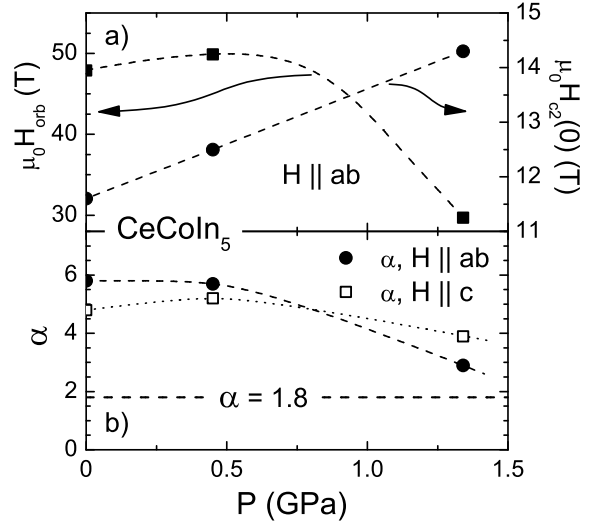


Fig. 4. Upper panel: left axis, orbital-limiting field, H_{orb} and, right axis, upper-critical field, $H_{c2}(0)$ versus pressure; lower panel: Maki-parameter, $\alpha = \sqrt{2}H_{orb}/H_P$, for $H \parallel ab$ and $H \parallel c$ as function of pressure.

tively. ℓ_{tr} increases substantially with increasing pressure. $\gamma_{2.8K} = C(2.8K)/T$ decreases only slightly, from about 350 mJ/mol K² at atmospheric pressure to 305 mJ/mol K² at 1.34 GPa. The main effect on ℓ_{tr} originates from the strong reduction of the resistivity in the normal state (see Fig. 3, upper panel), which is caused by a pressure-induced change of the inelastic scattering rate reflecting the reduction of the AFM-SF as expected for an increasing distance to the QCP.³¹

Pauli paramagnetism leads to an upper limit for the magnetic field which can be still supported by the superconductor, the Clogston paramagnetic limit $H_P = \Delta_0/\sqrt{2}\mu_B$ ³⁹ with Δ_0 the SC energy gap and μ_B the Bohr magneton. In addition, orbital effects also limit H_{c2} . Maki and Tsuneto⁹ showed that for a Pauli-limited system ($\alpha \geq 1$) the second order transition from the paramagnetic to the mixed state becomes instable, and for $\alpha \rightarrow \infty$ a first order transition is expected below $t_0 = T_0/T_c = 0.56$.⁹ The crossover from a second-order to a first-order phase transition observed at all pressures in CeCoIn₅ indicates that the strong Pauli-limiting effect is present also under pressure. The experimentally obtained t_0 is always smaller than the theoretical threshold $t_0 = T_0/T_c = 0.56$.

The Maki parameter $\alpha = \sqrt{2}H_{orb}/H_P$ can be estimated directly. The orbital-limiting field $H_{orb} = 0.7T_c(\partial H_{c2}(T)/\partial T)|_{T=T_c}$ ^{41,42} is easily accessible, while for the Pauli limiting field $H_P = H_c/(2\sqrt{\pi\chi_{spin}})$ the knowledge of the the spin susceptibility χ_{spin} is needed. H_c is the thermodynamic critical field. Without knowing χ_{spin} , we use a reasonable approximation $H_P = H_{c2}$. A more detailed analysis of the $H_{c2}(T)$ data within BCS theory gives values for H_P close to the approximation used here.^{43,44} The orbital-limiting field is strongly re-

duced on increasing the pressure, but is still about twice as large as $H_{c2}(0)$ at 1.34 GPa. This indicates that Pauli limiting still is the dominating effect in limiting the upper-critical field (see Fig. 4). The Maki parameter decreases for $H \parallel ab$ from $\alpha = 5.8$ at atmospheric pressure to $\alpha = 2.9$ at 1.34 GPa, which is still larger than the minimum value of $\alpha = 1.8$ required for the realization of the FFLO state in an s -wave superconductor.⁵

On applying pressure Pauli-limiting becomes weaker, but still exceeds the orbital-limiting effects. Furthermore, the crossover from a first-order to a second-order SC to normal phase transition at $H_0 = H_{c2}(T_0)$ is expected only in the case of a strongly Pauli limited upper critical field. From our observation that under pressure i) the Maki parameter $\alpha > 1.8$, ii) the ratio ℓ_{tr}/ξ_0 is increased, and iii) the anomaly at T_{FFLO} inside the SC state still exists, we conclude that the FFLO state is indeed formed in CeCoIn₅. However, the microscopic realization of the FFLO state in CeCoIn₅ needs further exploration. We found the FFLO phase expanding, although the Maki parameter decreases and while the spin fluctuations are strongly suppressed with increasing pressure. This suggests that SF have a strong detrimental effect on the FFLO phase, in agreement with theoretical predictions.⁴⁵ Recent NMR studies at atmospheric pressure suggest the existence of local moment magnetism inside the normal vortex cores,⁴⁶ in contrast to earlier reports.⁴⁷ This study highlights the intricate relation of magnetism and the FFLO phase close to $H_{c2}(0)$, consistent with the NFL behavior observed in the normal state close to $H_{c2}(0)$. Our data reveal that the expansion of the FFLO phase with increasing pressure is consistent with the strong suppression of the AFM-SF fluctuations.^{13,31} In fact LFL behavior is recovered at high pressure.^{13,31,34,35} In experiments carried out under pressure we would expect a drastic weakening of the spin polarization observed in the NMR measurements inside the FFLO phase at atmospheric pressure.

4. Conclusions

We have found that an anomaly at T_{FFLO} inside the SC phase in CeCoIn₅ close to $H_{c2}(0)$ established previously^{1,2,19–21} persists upon applying hydrostatic pressure. Also, the conditions for the formation of the FFLO state, namely Pauli-limiting exceeding orbital limiting and clean-limit SC, are found to be still fulfilled under pressure. While the Pauli-limiting effect is becoming weaker compared with orbital limiting on applying pressure, the FFLO phase is expanding, in contrast to what is naively expected. Considering the strong suppression of the AFM-SF on increasing pressure, the expansion of the FFLO phase can be explained by a strong detrimental effect of the AFM-SF on the FFLO state.

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